

Influence of chemical pressure effects on nonlinear thermal conductivity of intrinsically granular superconductors

Sergei Sergeenkov

*Departamento de Física, CCEN, Universidade Federal da Paraíba,
Cidade Universitária, 58051-970 João Pessoa, PB, Brazil*

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Abstract

Using a 2D model of capacitively coupled Josephson junction arrays (created by a network of twin boundary dislocations with strain fields acting as an insulating barrier between hole-rich domains in underdoped crystals), we study the influence of chemical pressure ($\nabla\mu$) on *nonlinear* (i.e., ∇T - dependent) thermal conductivity (NLTC) of an intrinsically granular superconductor. Quite a substantial enhancement of NLTC is predicted when intrinsic chemoelectric field $\mathbf{E}_\mu \propto \nabla\mu$ closely matches the externally produced thermoelectric field $\mathbf{E}_T \propto \nabla T$. The estimates of the model parameters suggest a realistic possibility to experimentally monitor this effect in non-stoichiometric high- T_C superconductors.

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High resolution imaging of the granular structure in underdoped $Bi_2Sr_2CaCu_2O_{8+\delta}$ crystals [1] has recently revealed an apparent segregation of its electronic structure into superconducting domains (of the order of a few nanometers) located in an electronically distinct background. In particular, it was found that at low levels of hole doping ($\delta < 0.2$), the holes become concentrated at certain hole-rich domains. (In this regard, it is interesting to mention a somewhat similar phenomenon of "chemical localization" that takes place in materials, composed of atoms of only metallic elements, exhibiting metal-insulator transitions [2].) Tunneling between such domains leads to intrinsic granular superconductivity (GS) in high- T_c superconductors (HTS). Probably one of the first examples of GS was observed in $YBa_2Cu_3O_{7-\delta}$ single crystals in the form of the so-called "fishtail" anomaly of magnetization [3]. The granular behavior has been related to the 2D clusters of oxygen defects forming twin boundaries (TBs) or dislocation walls within CuO plane that restrict supercurrent flow and allow excess flux to enter the crystal. Indeed, there are serious arguments to consider the TB in HTS as insulating regions of the Josephson SIS-type structure. Besides, a destruction of bulk superconductivity in these non-stoichiometric materials with increasing the oxygen deficiency parameter δ was found to follow a classical percolation theory [4]. In addition to their importance for understanding the underlying microscopic mechanisms governing HTS materials, the above experiments can provide rather versatile tools for designing chemically-controlled atomic scale Josephson junctions (JJs) and their arrays (JJAs) with pre-selected properties needed for manufacturing the modern quantum devices [5, 6]. To understand how GS manifests itself in non-stoichiometric crystals, let us invoke an analogy with the previously discussed dislocation models of grain-boundary Josephson junctions (GBJJs) (see, e.g., [7, 8] and further references therein). Recall that under plastic deformation, grain boundaries (GBs) (which are the natural sources of weak links in HTS), move rather rapidly via the movement of the grain boundary dislocations (GBDs) comprising these GBs. At the same time, observed [1, 3, 9, 10, 11] in HTS single crystals regular 2D dislocation networks of oxygen depleted regions (generated by the dissociation of $\langle 110 \rangle$ twinning dislocations) with the size d_0 of a few Burgers vectors, forming a triangular lattice with a spacing $d \geq d_0$ ranging from $10nm$ to $100nm$, can provide quite a realistic possibility for existence of 2D Josephson network within CuO plane. Recall furthermore that in a d -wave orthorhombic $YBCO$ crystal TBs are represented by tetragonal regions (in which all dislocations are equally spaced by d_0 and have the same Burgers

vector \mathbf{a} parallel to y -axis within CuO plane) which produce screened strain fields [10] $\epsilon(\mathbf{x}) = \epsilon(0)e^{-|\mathbf{x}|/d_0}$ with $|\mathbf{x}| = \sqrt{x^2 + y^2}$. Though in $YBa_2Cu_3O_{7-\delta}$ the ordinary oxygen diffusion $D = D_0e^{-U_d/k_BT}$ is extremely slow even near T_c (due to a rather high value of the activation energy U_d in these materials, typically $U_d \simeq 1\text{eV}$), in underdoped crystals (with oxygen-induced dislocations) there is a real possibility to facilitate oxygen transport via the so-called osmotic (pumping) mechanism [12, 13] which relates a local value of the chemical potential (chemical pressure) $\mu(\mathbf{x}) = \mu(0) + \nabla\mu \cdot \mathbf{x}$ with a local concentration of point defects as follows $c(\mathbf{x}) = e^{-\mu(\mathbf{x})/k_BT}$. Indeed, when in such a crystal there exists a nonequilibrium concentration of vacancies, dislocation is moved for atomic distance a by adding excess vacancies to the extraplane edge. The produced work is simply equal to the chemical potential of added vacancies. What is important, this mechanism allows us to explicitly incorporate the oxygen deficiency parameter δ into our model by relating it to the excess oxygen concentration of vacancies $c_v \equiv c(0)$ as follows $\delta = 1 - c_v$. As a result, the chemical potential of the single vacancy reads $\mu_v \equiv \mu(0) = -k_BT \log(1 - \delta) \simeq k_BT\delta$. Remarkably, the same osmotic mechanism was used by Gurevich and Pashitskii [10] to discuss the modification of oxygen vacancies concentration in the presence of the TB strain field. In particular, they argue that the change of $\epsilon(\mathbf{x})$ under an applied or chemically induced pressure results in a significant oxygen redistribution producing a highly inhomogeneous filamentary structure of oxygen-deficient nonsuperconducting regions along GB [11] (for underdoped superconductors, the vacancies tend to concentrate in the regions of compressed material). Hence, assuming the following connection between the variation of mechanical and chemical properties of planar defects, namely $\mu(\mathbf{x}) = K\Omega_0\epsilon(\mathbf{x})$ (where Ω_0 is an effective atomic volume of the vacancy and K is the bulk elastic modulus), we can study the properties of TB induced JJs under intrinsic chemical pressure $\nabla\mu$ (created by the variation of the oxygen doping parameter δ). More specifically, a single *SIS* type junction (comprising a Josephson network) is formed around TB due to a local depression of the superconducting order parameter $\Delta(\mathbf{x}) \propto \epsilon(\mathbf{x})$ over distance d_0 producing thus a weak link with (oxygen deficiency δ dependent) Josephson coupling $J(\delta) = \epsilon(\mathbf{x})J_0 = J_0(\delta)e^{-|\mathbf{x}|/d_0}$ where $J_0(\delta) = \epsilon(0)J_0 = (\mu_v/K\Omega_0)J_0$. Thus, the considered here model indeed describes chemically induced GS in underdoped systems (with $\delta \neq 0$) because, in accordance with the observations, for stoichiometric situation (when $\delta \simeq 0$), the Josephson coupling $J(\delta) \simeq 0$ and the system loses its explicitly granular signature.

There are several approaches for studying the thermal response of JJs and JJAs based on phenomenology of the Josephson effect in the presence of thermal gradients (see, e.g., [14, 15, 16, 17] and further references therein). In this paper, within a 2D model of capacitive JJAs (created by a regular 2D network of TB dislocations), we study the influence of chemical pressure effects (described by the gradient of the chemical potential $\nabla\mu$) on *nonlinear* (i.e. ∇T -dependent) thermal conductivity (NLTC) $\kappa(T, \nabla\mu; \nabla T)$ of intrinsically granular superconductors. As we shall see, in a sharp contrast with its linear counterpart, NLTC exhibits quite a pronounced enhancement when the intrinsically induced chemoelectric field $E_\mu = \frac{1}{2e}|\nabla\mu|$ matches the externally produced thermoelectric field $E_T = S_0|\nabla T|$.

To adequately describe transport properties of the chemically induced granular superconductor under a simultaneous influence of intrinsic chemical pressure $\nabla\mu(\mathbf{x}) = K\Omega_0\nabla\epsilon(\mathbf{x})$ and applied thermal gradient ∇T , we employ a model of 2D overdamped Josephson junction array which is based on the following total Hamiltonian [12]

$$\mathcal{H}(t) = \mathcal{H}_T(t) + \mathcal{H}_C(t), \quad (1)$$

where

$$\mathcal{H}_T(t) = \sum_{ij}^N J_{ij}[1 - \cos \phi_{ij}(t)] \quad (2)$$

is the well-known tunneling Hamiltonian, and

$$\mathcal{H}_C(t) = \sum_{ij}^N \frac{q_i(t)q_j(t)}{2C_{ij}} \quad (3)$$

accounts for a Coulomb contribution due to mutual capacitances C_{ij} between grains with a net charge

$$q_i(t) = \sum_{j=1}^N \int_0^t dt' I_{ij}(t') \quad (4)$$

induced by the normal current $I_{ij}(t) = V_{ij}(t)/R_{ij}$ within the array, where $V_{ij}(t) = (\frac{\Phi_0}{2\pi})\frac{\partial\phi_{ij}}{\partial t}$ and R_{ij} are the corresponding voltage and resistance between grains in their normal state.

According to the above-mentioned scenario, the tunneling Hamiltonian $\mathcal{H}_T(t)$ introduces a short-range (nearest-neighbor) interaction between N junctions (which are formed around oxygen-rich superconducting areas with phases $\phi_i(t)$), arranged in a two-dimensional (2D) lattice with coordinates $\mathbf{x}_i = (x_i, y_i)$. The areas are separated by oxygen-poor insulating boundaries (created by TB strain fields $\epsilon(\mathbf{x}_{ij})$) producing a short-range Josephson coupling $J_{ij} = J_0(\delta)e^{-|\mathbf{x}_{ij}|/d}$. Besides, as we have seen, the strain fields $\epsilon(\mathbf{x}_{ij})$ also control the dielectric

properties of the array. Namely, the mutual capacitances between grains are defined as $C_{ij} = 2\pi\epsilon_0\epsilon(\mathbf{x}_{ij})|\mathbf{x}_{ij}| = C_0(|\mathbf{x}_{ij}|/d)e^{-|\mathbf{x}_{ij}|/d}$. Thus, both the Josephson energy and the charging energy of the array vary exponentially with the distance $\mathbf{x}_{ij} = \mathbf{x}_i - \mathbf{x}_j$ between neighboring junctions (with d being an average grain size). The temperature dependence of Josephson coupling is governed by the standard expression [12]

$$J_{ij}(T) = J_{ij}(0) \left[\frac{\Delta(T)}{\Delta(0)} \right] \tanh \left[\frac{\Delta(T)}{2k_B T} \right] \quad (5)$$

where $J_{ij}(0) = [\Delta(0)/2](R_0/R_{ij})$ with $\Delta(T)$ being the temperature dependent gap parameter; $R_0 = h/4e^2$ and $R_{ij} \propto J_{ij}^{-1} = R_n e^{|\mathbf{x}_{ij}|/d}$ stand for quantum and normal resistance, respectively. For the explicit temperature dependence of the gap parameter we used the analytical approximation [12], namely $\Delta(T) = \Delta(0) \tanh \left(2.2 \sqrt{\frac{T_C - T}{T}} \right)$ which is valid for all temperatures.

By analogy with a constant electric field \mathbf{E} , a thermal gradient ∇T applied to a chemically induced JJA will cause a time evolution of the phase difference across insulating barriers as follows [15, 17, 18]

$$\phi_{ij}(t) = \phi_{ij}^0 + \frac{2e(\mathbf{E}_\mu - \mathbf{E}_T)\mathbf{x}_{ij}}{\hbar} t \quad (6)$$

Here ϕ_{ij}^0 is the initial phase difference (see below), $\mathbf{E}_\mu = \frac{1}{2e}\nabla\mu$ and $\mathbf{E}_T = S_0\nabla T$ are the induced chemoelectric and thermoelectric fields, respectively. S_0 is the Seebeck coefficient. In what follows, we assume, for simplicity, that $\nabla\mu = (\nabla_x\mu, 0, 0)$ with $\nabla_x\mu = \Delta\mu/d$, and that $\nabla T = (\nabla_x T, 0, 0)$.

In order to study the most interesting situation when the externally produced thermoelectric field \mathbf{E}_T becomes comparable with intrinsically induced chemoelectric field \mathbf{E}_μ , let us consider a *nonlinear* generalization of the conventional Fourier law and the resulting *nonlinear* thermal conductivity (NLTC) under the influence of chemical pressure. In what follows, by the NLTC we understand a ∇T -dependent thermal conductivity which is defined as follows [17]

$$\kappa(T, \nabla\mu; \nabla T) \equiv -\frac{1}{V} \left[\frac{\partial \langle Q_x \rangle}{\partial (\nabla_x T)} \right]_{\nabla T \neq 0} \quad (7)$$

where

$$\langle Q_x \rangle = \frac{1}{\tau} \int_0^\tau dt Q_x(t) \quad (8)$$

with $Q_x(t)$ being the longitudinal component of the total thermal flux which is defined (in a q-space representation) via the total energy conservation law as follows (V is sample's

volume, and τ is a characteristic time of the problem, see below)

$$\mathbf{Q}(t) \equiv \lim_{\mathbf{q} \rightarrow 0} \left[i \frac{\mathbf{q}}{q^2} \dot{\mathcal{H}}_{\mathbf{q}}(t) \right] \quad (9)$$

with

$$\dot{\mathcal{H}}_{\mathbf{q}} = \frac{1}{s} \int d^2x e^{i\mathbf{q}\mathbf{x}} \frac{\partial \mathcal{H}(\mathbf{x}, t)}{\partial t}, \quad (10)$$

Here, $s = 2\pi d^2$ is properly defined normalization area, and we made a usual substitution [17] $\frac{1}{N} \sum_{ij} A_{ij}(t) \rightarrow \frac{1}{s} \int d^2x A(\mathbf{x}, t)$ valid in the long-wavelength approximation ($\mathbf{q} \rightarrow 0$).

In view of Eqs.(1)-(10) and assuming $\phi_{ij}^0 = \pi/2$ for the initial phase difference (which maximizes the Josephson current of the network), we arrive at the following analytical expression for the temperature and chemical pressure dependence of the *nonlinear* thermal conductivity (TC) of a model granular superconductor

$$\kappa(T, \nabla\mu; \nabla T) = \kappa_n \left[1 + \beta_C(T) \frac{(1 + 4\eta^2)}{(1 + \eta^2)^4} \right] \quad (11)$$

Here, $\kappa_n \equiv \kappa(T_C, \nabla\mu; \nabla T) = 2\pi N d^2 S_0 I_C(0)/V \beta_C(0)$ is the normal state value of the NLTC, $\beta_C(T) = 2\pi I_C(T) C_0 R_n^2 / \Phi_0$ is the so-called Stewart-McCumber parameter [19] responsible for dissipative properties of the array with $I_c(T) = (2e/\hbar)J(T)$ being the critical current, and $\eta = (E_\mu - E_T)/E_0$ with $E_0 = \hbar/2ed\tau$ being a characteristic field. Notice that, as expected, in the limit $E_T \rightarrow 0$ (or when $\nabla T = 0$ in the rhs of Eq.(7)), Eq.(11) reduces to the expression for the linear thermal conductivity $\kappa(T, \nabla\mu; 0)$.

Figure 1 shows the dependence of the normalized NLTC $\kappa(T, \nabla\mu; \nabla T)/\kappa(T, 0; \nabla T)$ on reduced temperature T/T_C for chemical pressure $\Delta\mu/\Delta\mu_0 = 1$ (with $\Delta\mu_0 = \hbar/\tau$) for different values of the applied thermal gradient $\eta_T = S_T |\nabla T|/E_0$ with $\beta_C(0) = 1$ and $R_n = R_0$. Notice a markedly different behavior of the nonlinear TC (corresponding to $\eta_T = 0.4, 0.8$, and 1.2). Unlike its linear counterpart (shown by the upper curve with $\eta_T = 0$), it increases with increasing the temperature. Even more drastic difference between the linear and nonlinear TC can be seen in their chemical pressure dependence. Figure 2 depicts the behavior of the NLTC $\kappa(T, \nabla\mu; \nabla T)$ as a function of $\Delta\mu/\Delta\mu_0$ for different values of the dimensionless parameter $\eta_T = E_T/E_0$ and for $T = 0.2T_C$ (the other parameters are the same as before). As is clearly seen from this picture, in a sharp contrast with the pressure behavior of the linear TC (shown by the curve at the bottom and corresponding to $\eta_T = 0$), its nonlinear analog evolves with the chemoelectric field quite differently. Namely, NLTC exhibits chemically stimulated enhancement with a pronounced maximum at $E_\mu = \Delta\mu/2ed \simeq E_T$. To complete

our study, let us estimate the order of magnitude of the main model parameters. Starting with chemoelectric fields E_μ needed to observe the above-predicted nonlinear field effects in granular superconductors, we notice that according to Figure 2, the most interesting behavior of NLTC takes place for $E_\mu \simeq E_0$. Using typical *YBCO* parameters [10], $\epsilon(0) = 0.01$, $\Omega_0 = a_0^3$ with $a_0 = 0.2nm$, and $K = 115GPa$, we have $\mu_v = \epsilon(0)K\Omega_0 \simeq 1meV$ for an estimate of the chemical potential in HTS crystals, which defines the characteristic time $\tau \simeq \hbar/\mu_v \simeq 5 \times 10^{-11}s$. Furthermore, taking $d \simeq 10nm$ for typical values of the average grain size (created by oxygen-rich superconducting regions), we get $E_0 = \hbar/2ed\tau \simeq 5 \times 10^5 V/m$ and $|\nabla\mu| = \Delta\mu/d \simeq \mu_v/d \simeq 10^6 eV/m$ for the estimates of the characteristic field and chemical potential gradient (intrinsic chemical pressure), respectively. On the other hand, the maximum of NLTC occurs when this field nearly perfectly matches an "intrinsic" thermoelectric field $E_T = S_0|\nabla T|$ induced by an applied thermal gradient, that is when $E_\mu \simeq E_0 \simeq E_T$. Using [20] $S_0 \simeq 0.5\mu V/K$ for an estimate of the linear Seebeck coefficient in *YBCO*, we obtain $|\nabla T| \simeq E_0/S_0 \simeq 2 \times 10^6 K/m$ for the characteristic value of applied thermal gradient needed to observe the predicted here effects. Let us estimate now the absolute value of the normal state thermal conductivity $\kappa_n = 2\pi Nd^2 S_0 I_C(0)/V\beta_C(0)$. Recall that within the present scenario, the scattering of normal electrons is governed by the Stewart-McCumber parameter $\beta_C(T) = 2\pi I_C(T)C_0 R_n^2/\Phi_0$ due to the presence of the normal resistance R_n and mutual capacitance C_0 between the adjacent grains. The latter is estimated to be $C_0 \simeq 1aF$ using $d = 10nm$ for an average "grain" size. Furthermore, the critical current $I_C(0)$ can be estimated via the critical temperature T_C as follows, $I_C(0) \simeq 2\pi k_B T_C/\Phi_0$ which gives $I_C(0) \simeq 10\mu A$ (for $T_C \simeq 90K$) and leads to $\beta_C(0) \simeq 3$ for the value of the Stewart-McCumber parameter assuming $R_n \simeq R_0$ for the normal resistance which, in turn, results in $q \simeq \Phi_0/R_n \simeq 10^{-19}C$ and $E_C = q^2/2C_0 \simeq 0.1eV$ for an estimate of the "grain" charge and the Coulomb energy. Finally, assuming $V \simeq Nd^3$ for the sample's volume, and using the above-mentioned expressions for S_0 and $\beta_C(0)$, we obtain $\kappa_n \simeq 10^{-3}W/mK$ for an estimate of the maximum of the NLTC which is actually much higher than a similar estimate obtained for inductance controlled κ_n in electric-field driven NLTC [17], suggesting thus quite a realistic possibility to observe the predicted here non-trivial behavior of the thermal conductivity in non-stoichiometric high- T_C superconductors.

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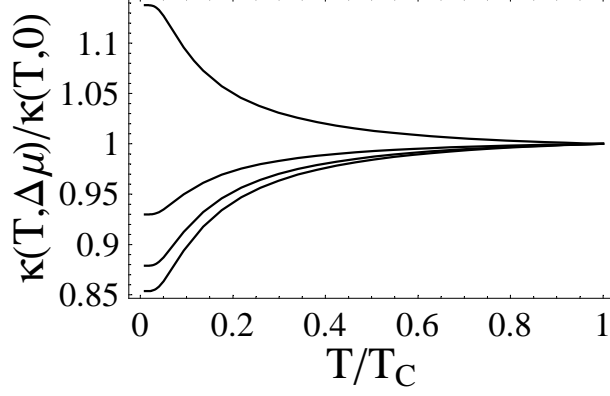


FIG. 1: The dependence of the *nonlinear* thermal conductivity on reduced temperature T/T_C for chemical pressure $\Delta\mu/\Delta\mu_0 = 1.0$ and for different values of the applied thermal gradient $\eta_T = S_0|\nabla T|/E_0$ ($\eta_T = 0, 0.4, 0.8$, and 1.2 , increasing from top to bottom), according to Eq.(11).

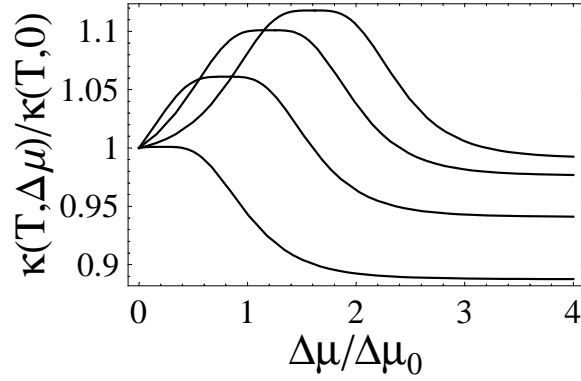


FIG. 2: The dependence of the *nonlinear* thermal conductivity on the chemical pressure $\Delta\mu/\Delta\mu_0$ for different values of the applied thermal gradient $\eta_T = S_0|\nabla T|/E_0$ ($\eta_T = 0, 0.4, 0.8$, and 1.2 , increasing from bottom to top), according to Eq.(11).